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AN EXPERIMENTAL INVESTIGATION OF THE
ALPHA-ELECTRON INTERACTION ACCOMPANYING
ALPHA DECAY

Thesis for the Degree of M. S.
MICHIGAN STATE UNIVERSITY

Allan R. McMillan

1960

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AN EXPERIMENTAL INVESTIGATION OF THE ALPHA-ELECTRON
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By

Allan R. McMillan

AN ABSTRACT

Submitted to the College of Science and Arts of Michigan
State University in partial fulfillment of the
requirements for the degree of

MASTER OF SCIENCE

Department of Physics

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Approved

William H. Miller

ABSTRACT

An experimental investigation of the alpha-electron interaction accompanying alpha decay was accomplished by examining the K and L x-rays accompanying the alpha decay of Po^{210} using NaI (Tl) scintillation detectors. The probability of the ejection of an electron from the atom was found by establishing the ratio of primary K and L shell vacancies to alpha particles emitted. The K shell ratio of $(2.75 \pm 0.25) \times 10^{-5}$ primary K shell vacancies per alpha particle emitted is, within experimental error, equal to the value predicted by Migdal. The L shell ratio of $(8.4) \times 10^{-4}$ primary L shell vacancies per alpha particle emitted is almost an order of magnitude larger than Migdal's theory predicts. Two experiments are suggested to establish the cause of this large difference.

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I. INTRODUCTION

Evidence of K and L shell ionization accompanying the alpha decay of Po^{210} has been advanced by Grace, Allen, West and Halban (1), Curie and Joliot (2), Barber and Helm (3), and Robinson and Bernstein (4). Grace et al. measured the number of K x-rays relative to the number of alpha particles and explained the K x-rays as the result of internal conversion of the 800 kev gamma ray accompanying the polonium decay. Barber and Helm also examined the number of K x-rays relative to the number of alpha particles and obtained results in agreement with Grace et al. However Barber and Helm concluded that the K x-rays were not due entirely to internal conversion, but rather to ionization caused by the emission of the alpha particles.

Curie and Joliot examined the lower energy L and M x-rays using an ionization chamber connected to an electroscope while Robinson and Bernstein investigated the L x-rays with proportional counters. In the course of these experiments Robinson and Bernstein demonstrated that the L x-rays are those of lead and not of polonium excited by alpha bombardment of Po^{210} atoms in the source.

Migdal (5) first considered this problem theoretically. Making use of the low velocity of the alpha particle compared to the velocity of the atomic electrons he calculated the probability of a primary electron vacancy in the final atom using time dependent adiabatic perturbation theory. In these calculations Migdal assumed that the alpha particle moves through the atom with a uniform velocity and used non-relativistic hydrogenic wave functions.

Barber and Helm obtained a ratio of primary K shell vacancies to the number of alpha particles emitted which was lower than the

ratio predicted by Migdal by a factor of about two. Robinson and Bernstein measured the ratio of L shell vacancies to alpha particles emitted and obtained results an order of magnitude larger than the value predicted by Migdal.

Levinger (6) re-examined the problem and introduced quadrupole terms in his calculations whereas Migdal had used only the dipole terms. Levinger's results predict a larger L shell yield than those of Migdal but still account for less than twenty per cent of the L shell x-ray yield observed by Robinson and Bernstein. Levinger's predictions for the K shell ionization are an order of magnitude smaller than the results of Migdal and an order of magnitude smaller than the experimental results of Barber and Helm.

Schwartz (7) has considered the problem in a different manner and has obtained results that are essentially equivalent to those of Migdal. Schwartz invokes the relative smallness of the charge of the alpha particle rather than its low velocity compared to the velocity of the atomic electrons. He takes into account the recoil of the nucleus and the change of charge that takes place at instant of alpha emission.

We are confronted with a large difference between the predictions of the theory and the results of the experiments. Since the most recent experiments were carried out in 1952 it was felt a new experiment with Po^{210} and perhaps some other alpha emitter might yield somewhat improved results with the use of better equipment.

II. DESCRIPTION OF EXPERIMENTS

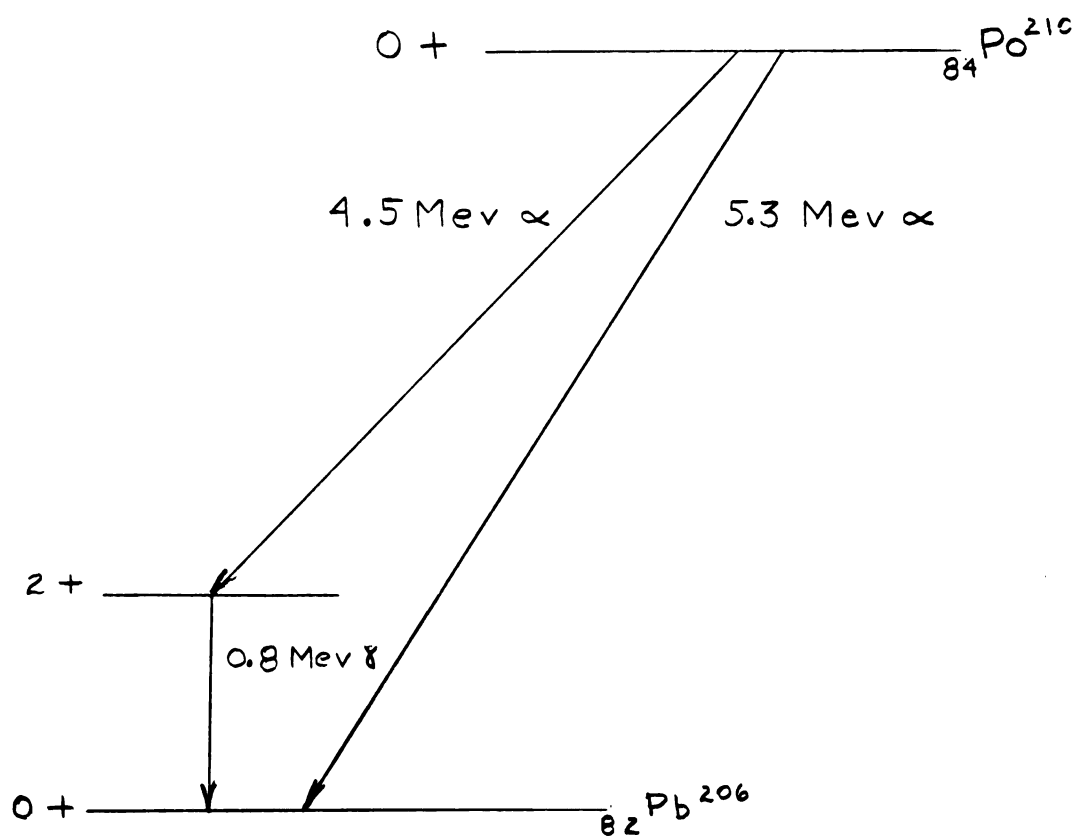
In the measurement of the ratio of K x-rays to alpha particles it is obvious that the use of the same source and geometry to count K x-rays and alphas would greatly simplify the problem. Unfortunately the low intensity of the K x-rays makes such a measurement almost impossible. The existence of an 800 kev gamma ray in the polonium decay scheme (fig. 1) makes possible an indirect measurement of the K x-ray to alpha particle ratio.

A 1 mc. source was prepared by evaporating to dryness a solution of Po^{210*} in the form of polonium nitrate on a film of mylar and attaching the mylar to a square of aluminum foil. Only the side of the source covered with the mylar was used for counting in the subsequent experiments. The source was mounted 3 cm above a 1 3/4" by 2" cylindrical NaI (Tl) crystal placed on an RCA 6342 photomultiplier tube. The photomultiplier was mounted on a cathode follower preamp and the entire counting arrangement was shielded on all sides with a layer of lead bricks. Pulses from the preamp were fed into an RIDL model A-61 amplifier and the amplified pulses were then analyzed and recorded by an RIDL model 3301 256 channel pulse height analyzer. In our experiment the spacing of the source above the detector is not critical because this distance enters the calculations only in the evaluation of the crystal efficiencies and essentially cancels out of the calculations since we are interested only in ratios.

The determination of the K x-ray to alpha ratio was accomplished by establishing the ratio of K x-rays to 800 kev gamma rays. This

* Obtained from Atomic Energy of Canada Ltd., Chalk River, Ontario.

Figure 1. Decay Scheme of Po^{210}



result is then combined with the ratio of 800 kev gamma ray to alpha particles as given by Ascoli et al. (8) to give an indirect determination of the K x-ray to alpha ratio.

The determination of the L x-ray to alpha ratio was obtained by establishing the ratio of L x-rays to K x-rays and combining the result with the experiment described above. The first of the L x-ray to K x-ray determinations was accomplished by placing the 1 mc source 4.2 mm above a 1 1/2" by 2 mm cylindrical NaI(Tl) crystal with a 4 mil beryllium window. A second experiment was performed with the same source 0.5 cm above an unmounted 1 1/2" by 1/4" cylindrical NaI(Tl) crystal. The electronic arrangement for these experiments was the same as described above.

The reasons for using the thin crystals were to reduce the background and Compton background from the higher-energy gamma and because the available thicker crystals were mounted in aluminum housings which attenuate the low energy L x-rays to a negligible intensity.

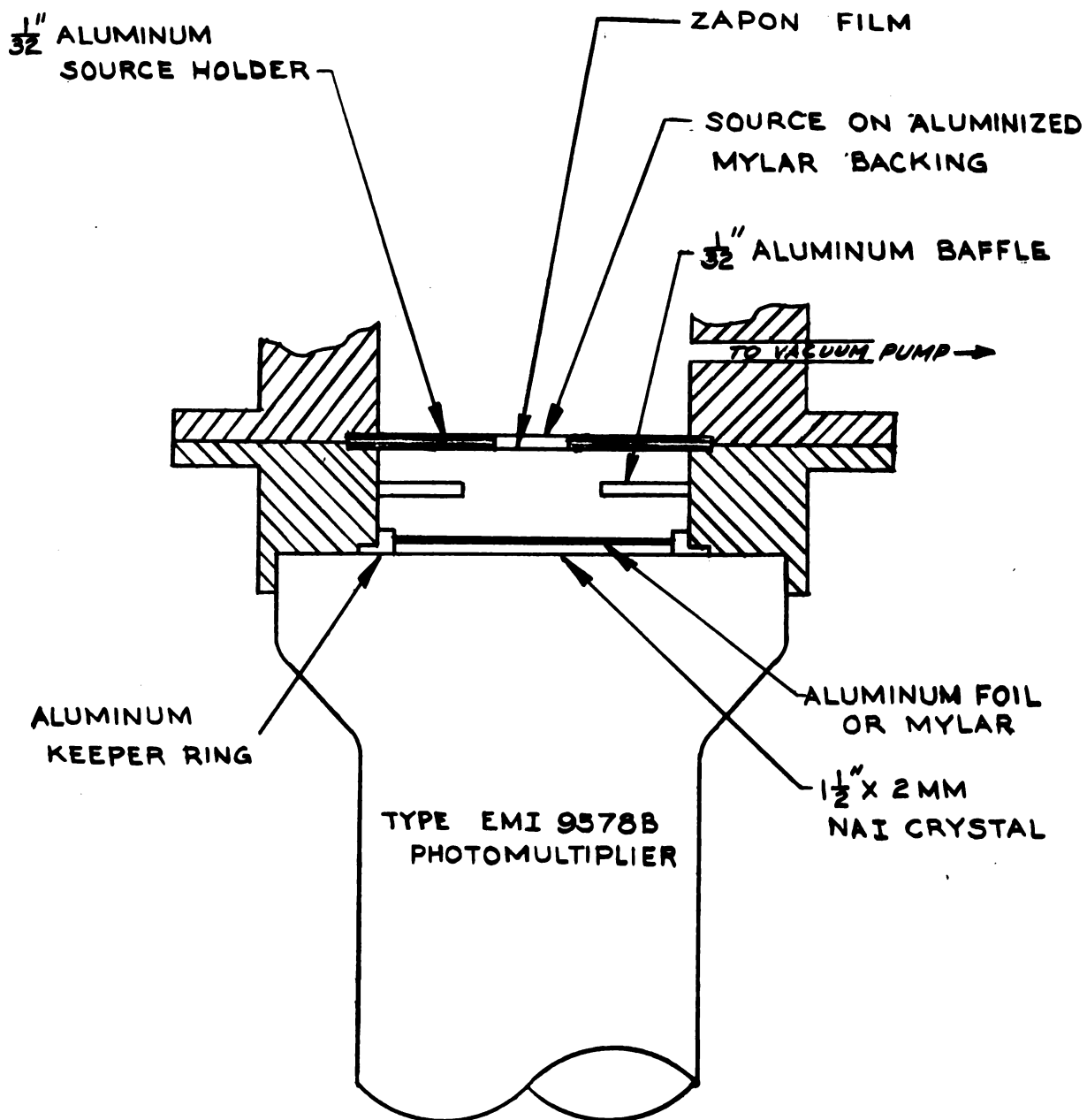
A direct determination of the L x-ray to alpha ratio was also performed. The difficulties of such an experiment are twofold. First, the use of a vacuum chamber is required for the alpha counting. The use of the vacuum chamber causes a great deal of scattering from the walls of the chamber and adds a low energy tail on the alpha peak (see fig. 7). This problem can be reduced by the use of baffles but we were unable to completely eradicate the problem. The second problem is that a source must be used which is strong enough that the L x-ray is well resolved from the background. Such a source will be so strong an alpha source that the detector must be partially shielded to prevent the alphas from saturating the amplifier and analyzer. The exposed area of the detector is critical in the determination of the efficiency of the detector. It is then very important to know the geometry of the

experiment as accurately as possible. In our experiments we knew the diameter of the detector to ± 0.0001 " and the height to ± 0.05 ".

The direct determination of the L x-ray to alpha ratio was performed using the small vacuum chamber shown in Fig. 2. A new source of ~ 0.1 mc was prepared by evaporating to dryness the polonium nitrate solution on a strip of aluminized mylar. The source was then centered over a $5/16$ " circular hole in a circular aluminum disk of thickness $1/32$ " and the other side of the hole was covered with a thin film of zapon. The purpose of the zapon film was to avoid contamination from recoiling clusters of Po^{210} . A $1\frac{1}{2}$ " by 2 mm unmounted NaI(Tl) crystal was placed on an EMI9578B photomultiplier and joined with the vacuum chamber. For the measurement of the alpha intensity it was necessary to use a small window over the face of the crystal to effectively reduce the size of the crystal. An aluminum plate with a $3/16$ " hole in the center was fitted over the face of the crystal. The aluminum plate was tapered such that the edge of the hole was reduced to a thickness of a few thousandths of an inch. This was done to reduce the alpha scattering from the edge of the window. For the measurements of the L x-ray intensity the plate was removed and replaced by a thick aluminum ring of inside diameter $1\frac{1}{4}$ " to insure accurate geometry.

A sheet of aluminized mylar of thickness $1.9 \pm 30\%$ mg/cm² was stretched over the crystal for the alpha counting to improve the optical qualities of the crystal. The mylar was replaced by a layer of aluminum foil of thickness 4.3 mg/cm² in the L x-ray measurement. The aluminum foil aided reflection in the crystal and stopped the alpha particles.

Figure 2. Small Vacuum Chamber



III. RESULTS

K X-ray to Gamma-ray Ratio Determination

The determination of K x-ray to gamma-ray ratio resulted in the spectra shown in Figs. 3 and 4. The dotted line in the gamma-ray spectrum shows the background with no source present. The dotted line in the K x-ray spectrum shows the assumed noise and Compton background.

The total number of quanta emitted by the source per unit time is obtained from the expression

$$N_o = \frac{N}{T(E) P(E) A_i t}$$

where N is the number of quanta detected under the peak

T(E) is the efficiency of the detector

P(E) is the peak to total ratio

A_i is any correction or attenuation factor

t is the duration of the measurement.

The NaI(Tl) efficiencies and peak to total ratios given by Heath et al. (9) are

T(81 kev)	0.0949 ± 0.0005
T(800 kev)	0.0370 ± 0.0003
P(81 kev)	1.000
P(800 kev)	0.342 ± 0.004

Figure 3. Po^{210} 800 Kev Gamma Ray Spectrum
100 Min. January 13.

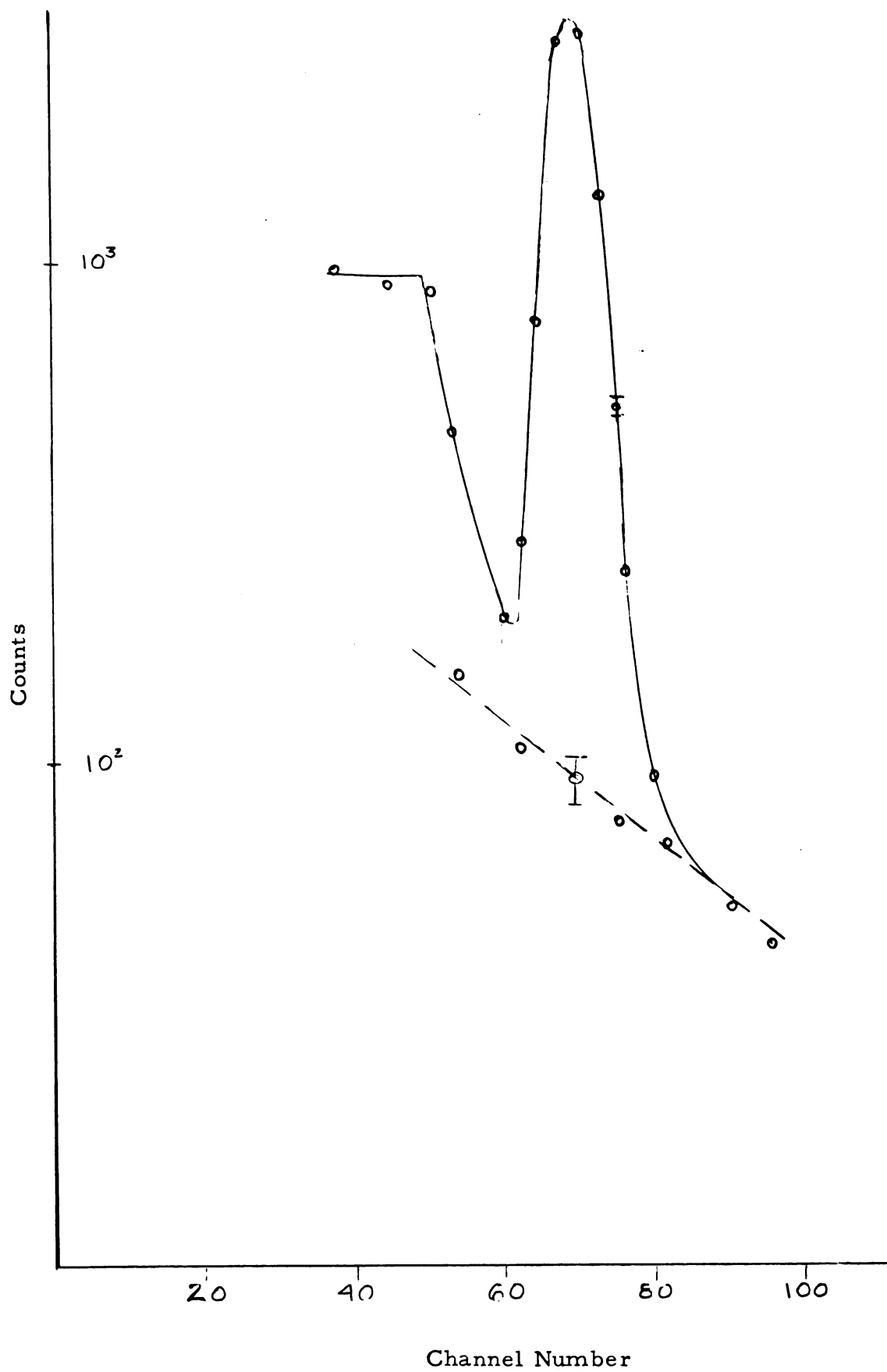
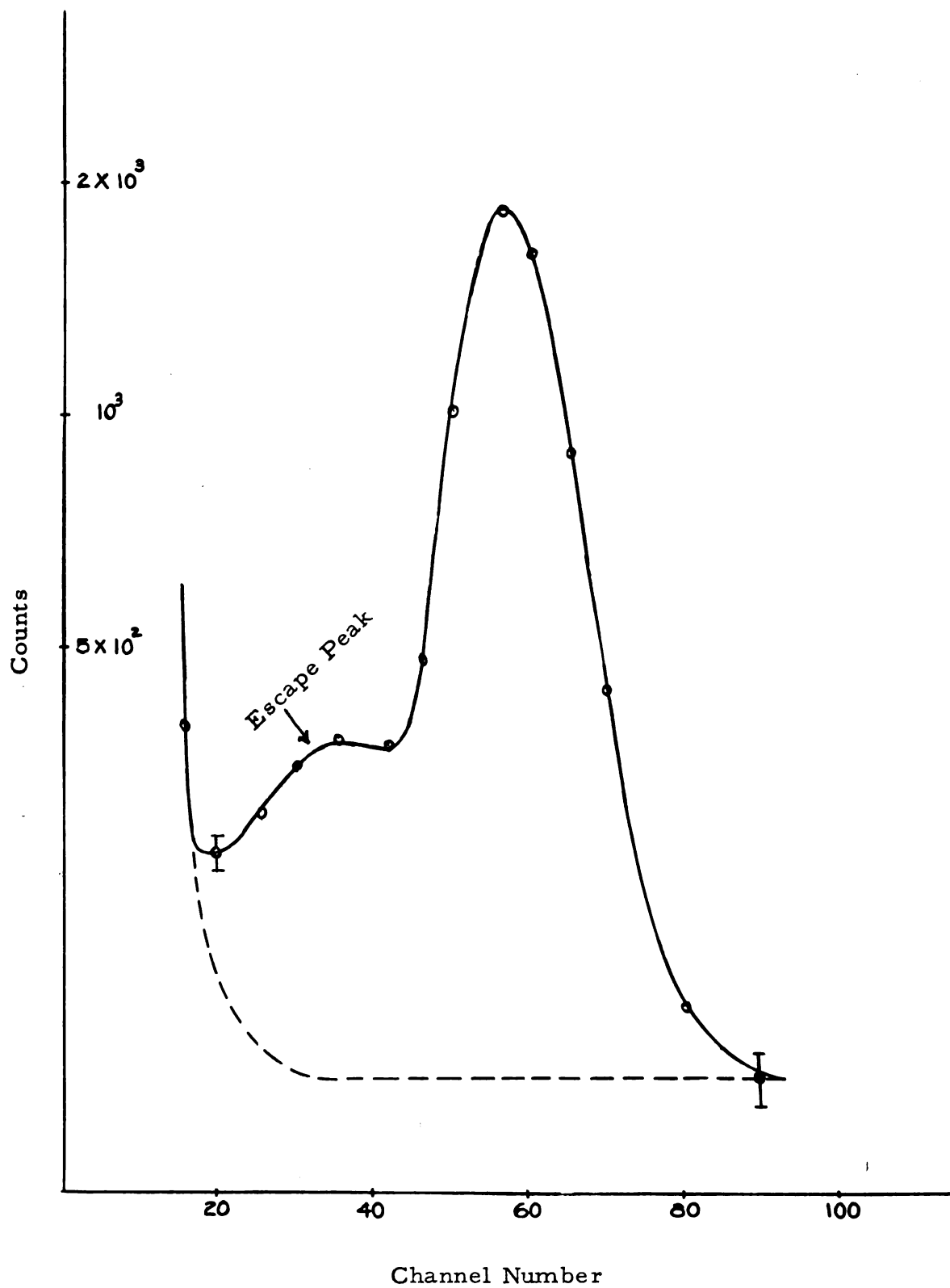


Figure 4. Po^{210} K X-ray Spectrum
100 Min. January 13.



The total number of counts under the K x-ray peak including the escape peak and excluding the background and Compton background* was $(33.3 \pm 0.24) \times 10^3$ per 100 minutes. The total number of counts under the 800 kev gamma ray peak excluding background was $(20.3 \pm 0.19) \times 10^3$ per 100 minutes. Then the ratio of the K x-rays to the 800 kev gamma rays is $0.218 \pm 0.005 (A_\gamma / A_k)$, where A_γ / A_k is the correction due to the relative absorption of the aluminum housing of the crystal mounting, calculated to be 1.016 ± 0.008 , and the fluorescence yield, 1.05 ± 0.01 (10). With these corrections the ratio of primary K shell vacancies to gamma rays is

$$\frac{N_k}{N_\gamma} = 0.23 \pm 0.01$$

Using calculated internal conversion coefficients (11) and the gamma ray to alpha particle ratio determined by Ascole et al. (8) the final value of primary K shell vacancies caused by the alpha emission per alpha particle emitted is determined to be

$$\frac{N_k}{N_\alpha} = (2.75 \pm 0.25) \times 10^{-6}$$

L X-Ray to K X-Ray Determination

The first determination of the L x-ray to K x-ray ratio was made using the 1 1/2" by 2 mm NaI crystal with the Be window. The result of this determination was

* An investigation of the Compton background was made using the 840 kev gamma ray of Mn⁵⁴ with the same experimental arrangement. This was found to be constant in the area corresponding to the K x-ray of lead.

$$\frac{N_L}{N_K} = 76 \pm 12$$

The main source of the large error in this experiment resulted from the evaluation of the detector efficiency using Simpson's rule for numerical integration and the uncertainty of the actual detector geometry. The numerical integration was found to be reliable to only 10% and the effective crystal size was less than the given diameter due to mounting material around the edge of the window. These errors were in different directions but would have been extremely difficult to evaluate.

In the subsequent experiments the L x-ray was not as well separated from the noise as in the experiment above. It was possible to use the L x-ray peak in the first experiment to analyze the peaks obtained in subsequent experiments. The shape and the resolution of the spectra in the later experiments were the same as those from the first experiment. We defined the peak position to be at 0.464 of the half maximum width measured from the low energy side of the line. Then the total number of counts under the peak was 2.316 ± 0.053 multiplied by the high energy portion of the peak, i.e., that portion of the peak above 0.464 of the half maximum width.

The second determination of the ratio of L x-rays to K x-rays was made using a $1\frac{1}{4}$ " by $\frac{1}{4}$ " NaI crystal as previously described. The K and L x-ray spectra obtained are shown in Figs 5 and 6. The determination of the shape of the K peak was made by fitting a Gaussian curve to the high energy edge of the L x-ray using the method of least squares and the Compton background was subtracted as in the K x-ray to gamma-ray determination. These corrections are shown in dotted lines in Fig. 6. The L x-ray was analyzed as outlined above.

Figure 5. Po^{210} L X-ray Spectrum
2 Min. Feb. 2

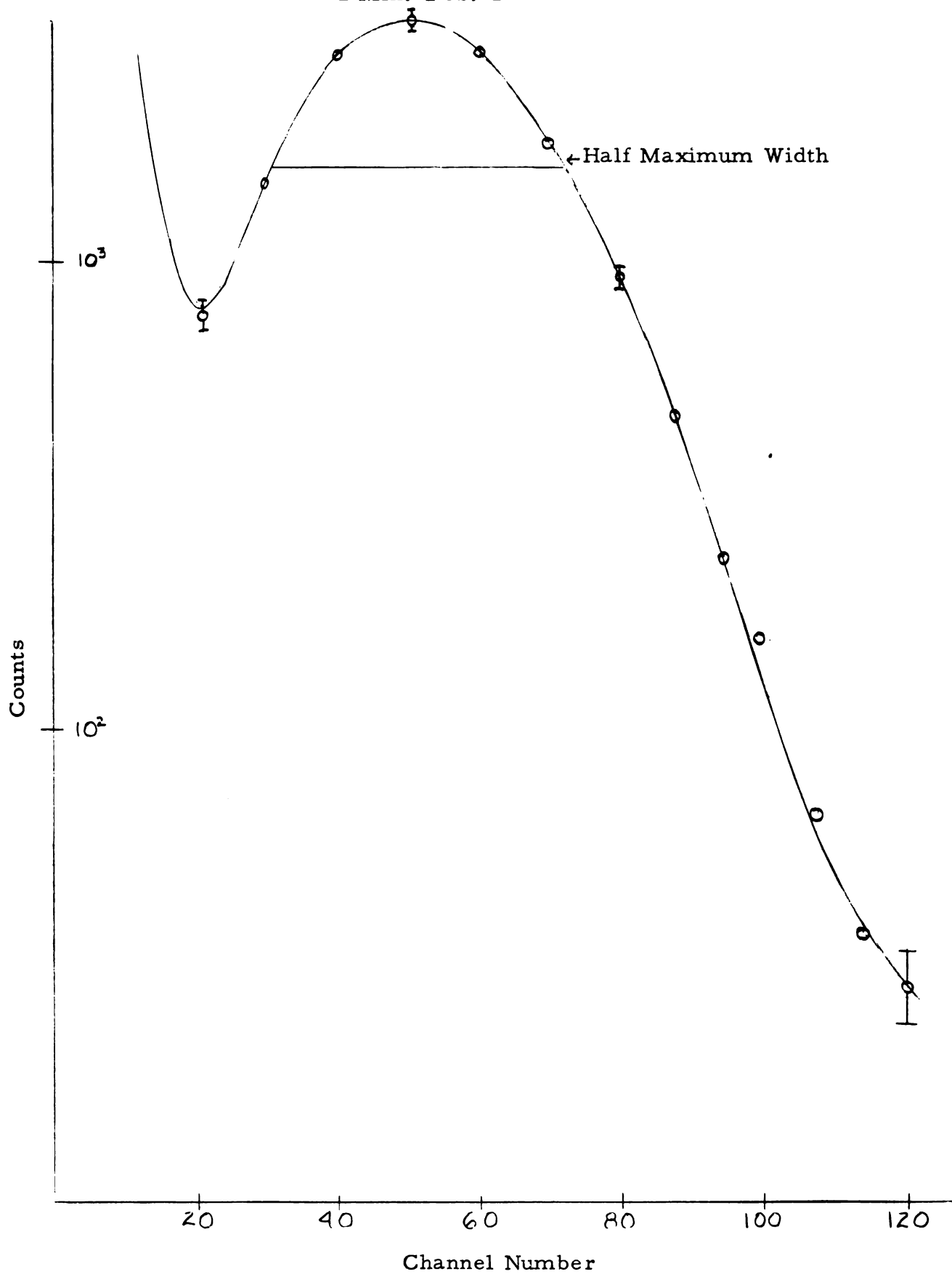
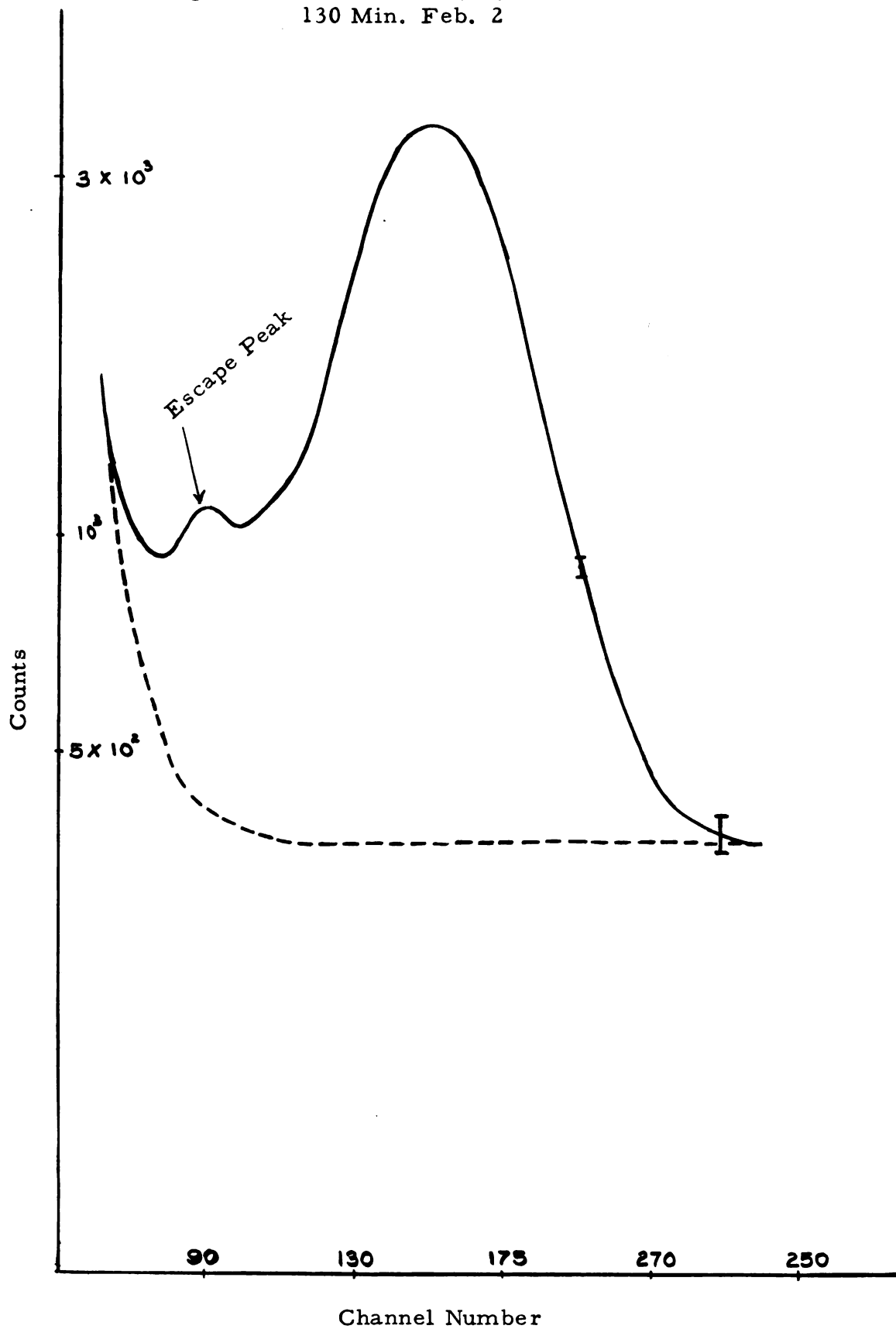


Figure 6. Po^{210} K X-ray Spectrum
130 Min. Feb. 2



The ratio of the detector efficiencies for the K and L x-rays was

$$\frac{T(80 \text{ kev})}{T(12 \text{ kev})} = 1.00 \pm 0.013$$

The ratio of the L x-rays to the K x-rays after correction for two layers of aluminum foil,* used to insure no alpha particles were counted, was

$$\frac{N_L}{N_K} = 79.0 \pm 2.1$$

Combining this result with the result of the K x-ray to alpha particle ratio we obtain

$$\frac{N_L}{N_\alpha} = (2.2 \pm 0.33) \times 10^{-4}$$

as the result of the indirect measurement of the ratio of L x-rays detected to the number of alpha particles emitted.

Results of the L to Alpha Experiment

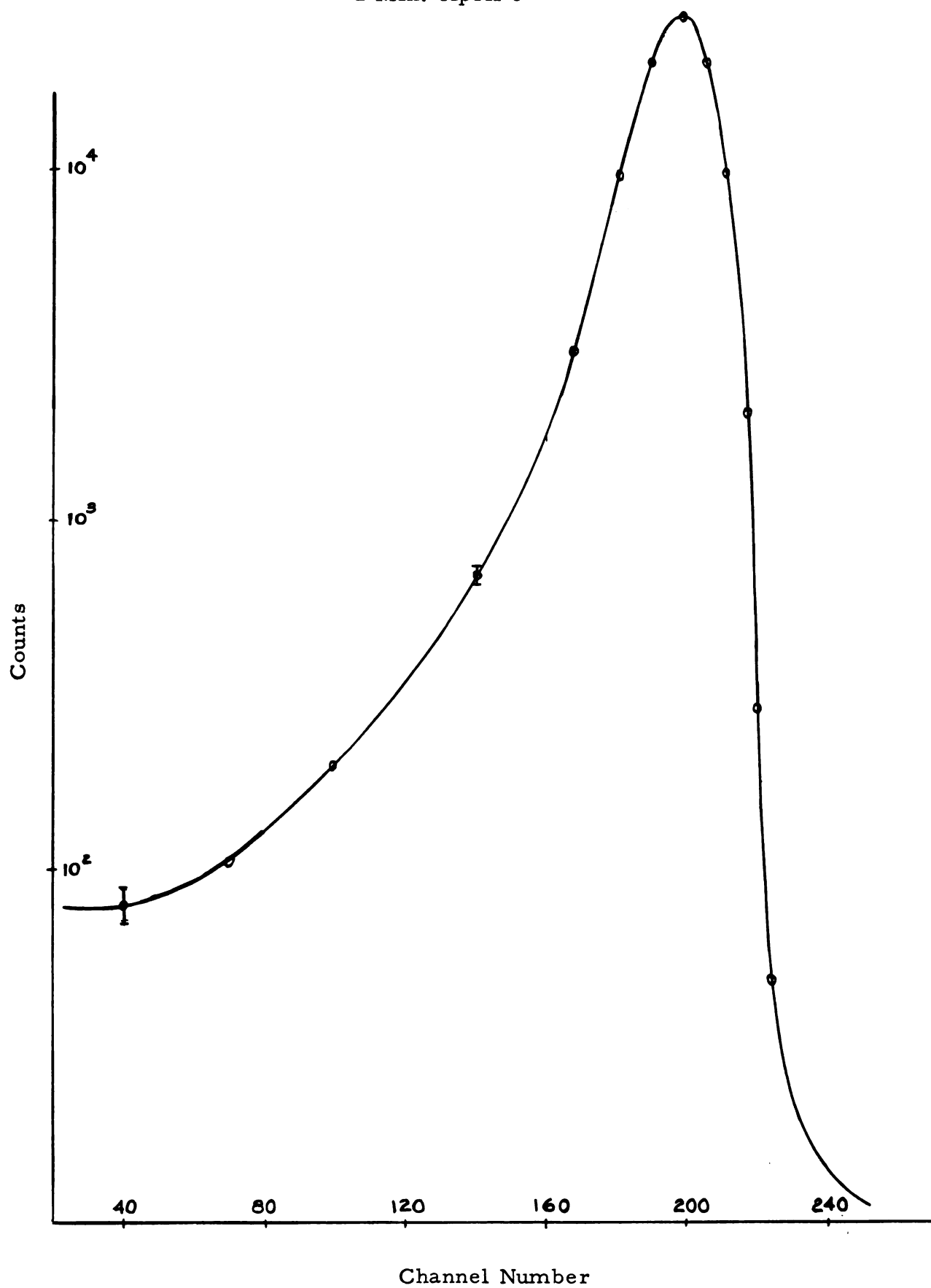
The determination of the ratio of the L x-ray to alpha particles by direct comparison of the L x-ray and alpha intensities was accomplished using the small vacuum chamber previously described. The alpha spectrum obtained is shown in Fig. 7. The L x-ray spectrum was essentially the same as that obtained in the previous experiment.

Determination of the NaI crystal efficiencies for these measurements consists merely of evaluating the solid angle subtended by the detector at the source since every alpha particle and L x-ray that

* Appendix.

Figure 7. Po^{210} Alpha Spectrum
2 Min. April 6

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strike the NaI crystal will contribute a count to the peak. The efficiencies of the detector corrected for the finite size of the source were

$$T(\alpha) = 0.0080 \pm 0.00056$$

$$T(L \text{ x-ray}) = 0.181 \pm 0.009$$

The L x-ray intensity was calculated as in the previous experiment and the alpha intensity was calculated by correcting the counts under the alpha peak for background, efficiency and time. The result was

$$\frac{N_L}{N_\alpha} = (2.49 \pm 0.23) \times 10^{-4} A_L$$

where A_L is the attenuation factor due to one layer of aluminum foil placed over the face of the NaI crystal to stop the alpha particles in the L x-ray measurement and equal to 1.12 ± 0.01 .^{*} Then the ratio of the number of L x-rays emitted by the source to the number of alpha particles emitted is

$$\frac{N_L}{N_\alpha} = (2.80 \pm 0.24) \times 10^{-4}$$

It will be noted that there is no definite cutoff of the alpha peak on the low energy end. This is due to scattering of alpha particles from the vacuum chamber and straggling. In the analysis of this peak we counted as the peak that region between channels thirty and 240. The area from channel thirty to channel ninety contributed less than one percent to the total area under the peak. The choice of channel thirty as the low energy end of the peak was arbitrary but does not appreciably affect the analysis of the peak.

^{*}Appendix.

IV. COMPARISON OF RESULTS AND DISCUSSION

Barber and Helm have previously obtained results for the K x-ray to alpha ratio by a direct measurement of the K x-ray and alpha intensities. A comparison of their results and our results is given below.

Barber and Helm

$$(K/\alpha) \qquad (2.2 \pm 0.42) \times 10^{-6}$$

Present experiments

$$(K/\alpha) \qquad (2.75 \pm 0.25) \times 10^{-6}$$

These ratios are the ratio of primary K shell vacancies to alpha particles emitted.

Barber and Helm have obtained a K x-ray spectrum that is very similar to the K x-ray spectra which we obtained (see Figs. 4 and 6). In our spectra the escape peak is evident. The spectrum of Barber and Helm gives no indication of a resolved escape peak although they made corrections for it. We feel that they have left out another portion of the peak that should be included. We have investigated the Compton background under the K peak and concluded that this background is very nearly constant with respect to energy in this energy region. Barber and Helm have apparently assumed that this Compton background is strictly decreasing under the K peak. We believe that they have excluded some of the K x-rays in this analysis and that their results are thus low by perhaps ten percent. With this correction their results are in fair agreement with ours.

The direct K to alpha determination that Barber and Helm carried out was accomplished by reducing the effective efficiency of

the detector by means of a shutter between the source and the detector similar to the method we used for the direct N_L/N determination. Although the diameter of the hole in their shutter is given as 0.033 cm no statement is made as to the accuracy with which the area was determined. As was mentioned before the efficiency of the detector is sensitive to the area of such a small aperture.

Rubinson and Bernstein have obtained results for the L x-ray to alpha ratio which differ somewhat from our indirect ratio obtained by combining the L to K ratio with the K to alpha ratio. Their ratio agrees to within experimental errors with the direct L x-ray to alpha ratio we have obtained. A comparison of the results of Rubinson and Bernstein and our results is given below.

Rubinson and Bernstein

$$(L/\alpha) \quad (2.93 \pm 0.44) \times 10^{-4}$$

Present experiments

$$(L/\alpha) \text{ indirectly} \quad (2.2 \pm 0.33) \times 10^{-4}$$

$$(L/\alpha) \text{ directly} \quad (2.8 \pm 0.24) \times 10^{-4}$$

Comparing our results with the results of Rubinson and Bernstein we believe the higher ratio to be the more accurate. The indirect ratio has been obtained by combining three ratios and treating the errors as random errors. The ratios given above are not corrected for fluorescence yield.

It is interesting to compare the above results with the theoretical predictions of Migdal (5) and Schwartz (7). The probability, W_{nl} , for ejection of an nl electron is, using the notation of Migdal

$$W_{nl} = (v/v_e)^2 C_{nl}/Z^2$$

where v is the velocity of the alpha particle

v_e is the velocity of the atomic electron,

C_{nl} is a constant that depends on n and l .

The C_{nl} have been calculated by Migdal and are given as

$$C_{10} = 2.2$$

$$C_{20} + C_{21} = 20.5$$

$$C_{30} + C_{31} + C_{32} = 68.6$$

Using this expression for the K electrons we obtain the result

$$W_K = 2.5 \times 10^{-6}$$

and for the L electrons

$$W_L = 1.13 \times 10^{-4}$$

In the calculation of the L probability there is some question as to what to use for Z the atomic number. Making use of the internal screening constants given by Sommerfeld and Wentzel (12) we have used $Z = 80$ for the 2, 0 electrons and $Z = 78.5$ for the 2, 1 electrons in obtaining the results given above.

The experimental result we have obtained for the K shell ionization agrees quite well with the theoretical result. The L shell results do not agree at all however. Using the values for the fluorescence yield for the L shell calculated by Kinnsey (13), we obtain a ratio of the primary L shell vacancies per alpha particle emitted of 8.4×10^{-4} . The theoretical result accounts for less than twenty percent of the observed result. An examination of the approximation used in the calculations of Migdal does not appear to yield the reason for the large difference in the predicted results and the observed results for the L shell. These approximations are:

- 1) The charge of the alpha particle is small compared to the charge of the daughter nucleus. For Po^{210} the ratio of the charge of the alpha particle to the charge of the daughter nucleus is 0.025.
- 2) The velocity of the alpha particle is much less than that of the electron. For the K shell electrons the ratio of the alpha velocity to the electron velocity is 0.1 and for the L shell electrons the ratio is 0.2.
- 3) The use of nonrelativistic hydrogenic wave functions. As mentioned by Robinson and Bernstein this approximation should be valid for the L shell electrons.

One possible solution to the problem of the large difference in the predicted L shell yield and the observed result could be the existence of a low lying level of the Pb^{206} . This level would have to have an energy less than that of the K shell binding energy and greater than that of the L shell in order that it would not affect the K shell yield and still be L shell converted. The gamma transition would have to be almost completely internally converted such that the gamma itself would not be observed. We have investigated the possibility of such an excited level. The 800 kev gamma cannot be involved to any appreciable extent because its intensity is a factor of ten lower than the effect we are attempting to explain. We conclude that the low lying level, if it exists, must be reached through an alpha transition from the Po^{210} .

We have examined the hinderance due to the difference in energy of the alpha that would be required for the above transition. Using the empirical formula from Perlman and Rasmussen (14) relating the half life to the disintegration energy of alpha decay

$$\log T_{\frac{1}{2}} = \frac{A}{\sqrt{Q}} + B$$

where $T_{\frac{1}{2}}$ is the half life in seconds

Q is the disintegration energy in Mev

Z is the charge of the parent nucleus

A and B are constants (For Po $A = 129.35$ $B = 49.9229$)

we find a hinderance due to the difference in energy of less than ten is expected. The spin of the state may also add to the hinderance. Blatt and Weisskopf (15) quote calculated hinderances due to spin which indicate that spins of less than 4 would give an effect that would be at least a hundred times too large. The higher spin states, ≥ 4 , are hindered enough and would be very highly converted in the L shell (11). It appears that a high spin, low energy excited state of lead could cause the large L shell yield. A spin of 4 would give rise to an E-4 transition which should have a lifetime of $\sim 10^9$ seconds (16). By chemically separating the lead from a sample of Po^{210} and examining the lead for L x-rays it would be possible to investigate the possibility of such a high spin, low energy state.

However lead is a magic number nucleus and such low lying states are not to be expected. An investigation of the even-even lead isotopes shows that none have an excited level of energy less than 570 kev. That a low energy excited level of Pb^{206} is causing the high L shell yield appears unlikely.

Further investigation of the L shell yield for other isotopes would aid in the solution of this problem. Unfortunately there are very few alpha emitters which are adapted to this experiment. The only other isotope which appears promising is Sm^{147} which is a pure alpha emitter. The long half life ($\sim 10^{11}$ years) and the low energy (~ 5 kev) of the L x-ray of Nd^{143} to which the samarium decays make

it very difficult to examine using the scintillation method. We have been able to observe what appears to be the L x-ray accompanying this decay but were not able to obtain accurate results due to a lack of resolution and a high noise level. We hope in the near future to obtain accurate results.

The theory of Migdal predicts for the samarium 5×10^{-4} primary L shell vacancies per alpha particle emitted. The L x-ray yield would be about 1/3 of the number of primary vacancies. An experimental result significantly larger than this predicted result would indicate that the theory is inadequate in its description of the process in the case of the L shell. A result in agreement with the predicted value would indicate that the decay scheme of Po^{210} is probably inaccurate.

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APPENDIX

In the course of these experiments it became necessary to evaluate the absorption of the L x-ray by the thin Al foil (4.3 mg/cm^2). The $1\frac{1}{2}$ " by 2 mm NaI (Tl) crystal with a 4 mil Be window mounted on an RCA 6342 photomultiplier tube was used to evaluate this absorption.

Spectra with one to seven layers of the Al foil between the source and the crystal were recorded and analyzed. A least squares fit of the logarithm of the L x-ray counts under the peak vs. the number of layers of Al foil was made with the result (Fig. 8)

$$\text{Log } N = 5.18836 - 0.04948x$$

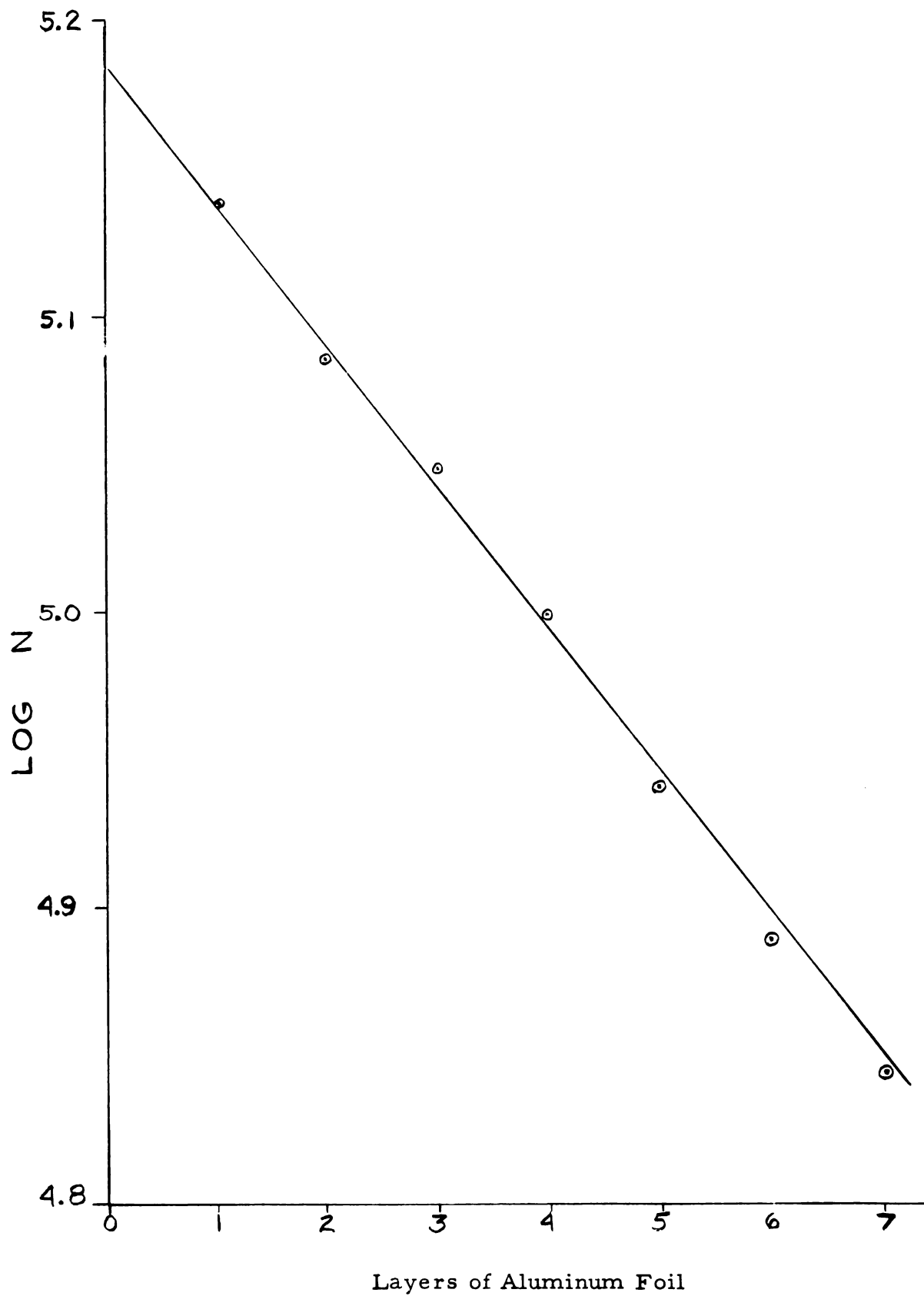
where x is the number of layers of Al foil.

A_1	1.12 ± 0.011	(one layer of Al foil)
A_2	1.26 ± 0.013	(two layers of Al foil)

There is no significant difference between having the Al foil adjacent to the source or the detector. The results of the experiment are usable in any situation where the geometry is similar to that of this experiment.

The absorption of the K x-ray due to one layer of Al foil was found to be negligible.

Figure 8. Log N vs. Layers of Al Foil



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